Odd-Parity Triplet Pair Induced by Hund's Rule Coupling

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(Dated: February 6, 2008)

We discuss microscopic aspects of odd-parity triplet pair in orbital degenerate systems. From the concept of off-diagonal long-range order, a pair state is unambiguously defined as the eigenstate with the maximum eigenvalue of pair correlation function. Performing this scheme by a numerical technique, we clarify that the odd-parity triplet pair occurs as an out-of-phase combination of local triplets induced by Hund's rule coupling for the lattice including two sites in the unit cell.

PACS numbers: 74.20.Rp, 74.20.Mn, 74.70.Tx, 74.70.Pq

Recently triplet superconductivity has been found in several compounds with strong electron correlation. A pioneering material should be UPt_3 [1], which has been considered to have spin-triplet pairing from experimental evidence for multi superconducting phases similar to the phase diagram of ^{3}He. The odd-parity triplet pair has been eventually confirmed by the experimental fact that the Knight shift does not change through the superconducting transition temperature T_c [2]. In other f-electron compounds, triplet superconductivity has been also suggested. For instance, coexistence of superconductivity and ferromagnetism has been discovered in UGe_2 [3] and URhGe [4]. It is naively believed that spin triplet pair appears in the ferromagnetic (FM) phase. Recently UNi_2Al_3 has been also considered as a triplet superconductor from NMR measurements [5].

Among transition metal oxides, Sr_2RuO_4 has attracted much attention since its discovery in 1994 [6]. It is confirmed that this material has spin triplet pair from NMR measurements [7], although it is isostructural to La_2CuO_4 which is the mother compound of high-T_c cuprates with singlet d-wave superconductivity. In the d-electron system such as $ZrZn_2$, it has been also reported that superconductivity appears in the FM phase [8] and spin-triplet pairing is expected to occur in this material.

Regarding the mechanism of triplet superconductivity, FM spin fluctuation was considered to mediate the triplet pair also in strongly correlated materials by analogy with superfluid ^{3}He. However, paramagnons are not always dominant in the spin fluctuation spectrum of those materials. In fact, in Sr_2RuO_4, significant enhancement of the incommensurate antiferromagnetic spin fluctuation has been observed in neutron scattering experiments [9]. Namely, in contrast to the naive expectation, paramagnons do not always play a central role in the occurrence of triplet superconductivity in the solid state. It is still a puzzling and challenging problem to clarify a key issue to determine the Cooper-pair symmetry.

One possible scenario is based on the Hund's rule coupling, which stabilizes the local triplet pair composed of a couple of electrons between different orbitals. Then, the triplet superconductivity is naively expected to occur, but in such a local picture, it is questionable whether the anisotropic Cooper-pair is stabilized or not. This point has casted a serious doubt on the scenario, but in the course of the investigation on UPt_3, it has been pointed out that an odd-parity triplet pair is realized only when the inversion center exists external to the f-shell ions [10]. This has been further examined by the detailed group-theoretical analysis [11] and the estimation of pairing potential with the use of band-calculation results [12]. However, the discussions have been in a phenomenological level and it is highly required that the triplet pair induced by the Hund's rule coupling should be investigated from the microscopic viewpoint.

In this Letter, we attempt to gain an insight into triplet pairing induced by Hund's rule coupling. First we reconsider the symmetry argument on triplet pair in the weak-coupling limit. It is again found that the odd-parity triplet pair induced by the Hund's rule interaction occurs only for the non Bravais lattice. In order to confirm this result from the microscopic viewpoint, we carefully analyze an orbital degenerate model on the two-dimensional (2D) square and honeycomb lattices. It is emphasized that the pair state is determined unambiguously by diagonalizing the pair correlation function based on the concept of off-diagonal long-range order [13]. Then, we can visualize the pair wavefunction in an unbiased manner, clearly indicating that the odd-parity triplet pair in the FM phase occurs as an out-of-phase combination of local triplets induced by Hund's rule coupling in the 2D honeycomb lattice with two sites in the unit cell.

Let us start our discussion on the Cooper-pair amplitude in the lattice with inversion symmetry. The pair operator composed of a couple of electrons is defined in the second quantized form as

$$\phi_{\mathbf{i}\mu\alpha\sigma,\,\mathbf{j}\nu\beta\tau} = c_{\mathbf{i}\mu\alpha\sigma}c_{\mathbf{j}\nu\beta\tau}, \tag{1}$$

where $c_{\mathbf{i}\mu\alpha\sigma}$ is an annihilation operator of an electron with spin σ in the orbital α on the site μ included in the unit cell $\mathbf{i}$. We consider the Fourier transform of $c_{\mathbf{i}\mu\alpha\sigma}$ as

$$c_{\mathbf{i}\mu\alpha\sigma} = \sum_{n,\boldsymbol{k}} e^{i\boldsymbol{k}\cdot\boldsymbol{R}_{\mathbf{i}}}\Lambda_{\mu\alpha}^{\boldsymbol{k}n}a_{\boldsymbol{k}n\sigma}, \tag{2}$$

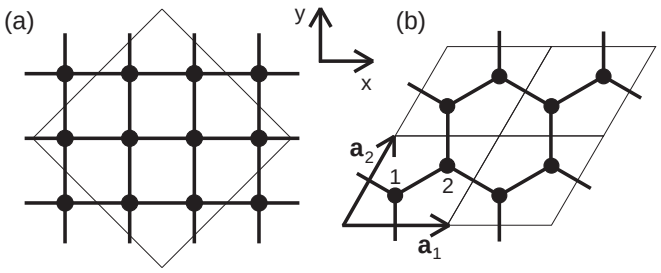

FIG. 1: (a) Two-dimensional square and (b) honeycomb lattices. Thin lines in (a) denotes 8-site cluster, while in (b), they indicate 8-site cluster composed of 4 unit cells including two sites, 1 and 2. Thick arrows define vectors $\mathbf{a}_1$ and $\mathbf{a}_2$.

where $a_{\boldsymbol{k}n\sigma}$ is an annihilation operator for an electron with momentum $\boldsymbol{k}$ and spin σ in the n-th band. Note that $\Lambda^{\boldsymbol{k}n}_{\mu\alpha}$ is the matrix element of the unitary transformation between orbitals and bands.

Now let us consider the Cooper pair of which the total momentum is equal to zero in the weak-coupling limit. Then, the Cooper-pair amplitude is given by

$$\langle\phi_{\mathbf{i}\mu\alpha\sigma,\,\mathbf{j}\nu\beta\tau}\rangle=\sum_{n,\boldsymbol{k}} e^{i\boldsymbol{k}\cdot(\boldsymbol{R}_\mathbf{i}-\boldsymbol{R}_\mathbf{j})}\Lambda^{\boldsymbol{k}n}_{\mu\alpha}\Lambda^{-\boldsymbol{k}n}_{\nu\beta}\langle a_{\boldsymbol{k}n\sigma}a_{-\boldsymbol{k}n\tau}\rangle. \quad (3)$$

Note that the Cooper pair is formed only by a couple of electrons on the same electronic band, since the pair composed of electrons in different bands is not stable in general in the weak-coupling limit.

Here we introduce an inversion operator $\mathcal{P}$, which acts generally on the electron operator as

$$\mathcal{P}c_{\mathbf{i}\mu\alpha\sigma}\mathcal{P}^{-1}=P_\alpha c_{\mathbf{i}\nu\alpha\sigma}, \quad (4)$$

where P_α denotes the parity for the inversion of atomic orbital, depending on the angular momentum. In this paper, we assume the pairing in the orbitals with the same angular momentum, indicating P_α=P_β.

For the Bravais lattice in which one site is included in the unit cell, typically the square lattice shown in Fig. 1(a), the inversion center is located on the site. Thus, we can simply suppress the indices μ and ν in this case. Combing Eqs. (2) and (4), we obtain

$$\Lambda^{\boldsymbol{k}n}_{\alpha}=P_\alpha\Lambda^{-\boldsymbol{k}n}_{\alpha}, \quad (5)$$

for the Bravais lattice. On the other hand, for the non Bravais lattice, μ is not equal to ν in Eq. (4). As a typical example, here we consider the honeycomb lattice composed of unit cells including two sites, as shown in Fig. 1(b). In this case, the inversion center is located at the center of two sites and we obtain

$$\Lambda^{\boldsymbol{k}n}_{1\alpha}=P_\alpha\Lambda^{-\boldsymbol{k}n}_{2\alpha}. \quad (6)$$

First we consider the Cooper pair in the Bravais lattice. Note again that the index μ is suppressed in this case.

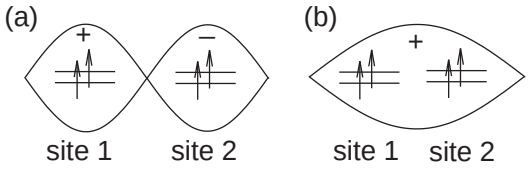

FIG. 2: Schematic views for wavefunctions of (a) odd-parity and (b) even-parity triplet pairs in the honeycomb lattice.

After algebraic calculations, spin singlet (S) and triplet (T) pairs are, respectively, given by

$$\langle\phi^{\mathrm{S}}_{\mathbf{i}\alpha,\,\mathbf{j}\beta}\rangle=\sum_{n,\boldsymbol{k}}\cos[\boldsymbol{k}\cdot(\boldsymbol{R}_\mathbf{i}-\boldsymbol{R}_\mathbf{j})]\Lambda^{-\boldsymbol{k}n}_{\alpha}\Lambda^{\boldsymbol{k}n}_{\beta}S_{\boldsymbol{k}n}, \quad (7)$$

and

$$\langle\phi^{\mathrm{T},z}_{\mathbf{i}\alpha,\,\mathbf{j}\beta}\rangle=\sum_{n,\boldsymbol{k}} i\sin[\boldsymbol{k}\cdot(\boldsymbol{R}_\mathbf{i}-\boldsymbol{R}_\mathbf{j})]\Lambda^{-\boldsymbol{k}n}_{\alpha}\Lambda^{\boldsymbol{k}n}_{\beta}T^{z}_{\boldsymbol{k}n}, \quad (8)$$

where $S_{\boldsymbol{k}n}=(\langle a_{\boldsymbol{k}n\uparrow}a_{-\boldsymbol{k}n\downarrow}\rangle-\langle a_{\boldsymbol{k}n\downarrow}a_{-\boldsymbol{k}n\uparrow}\rangle)/\sqrt{2}$, $T^{+1}_{\boldsymbol{k}n}=\langle a_{\boldsymbol{k}n\uparrow}a_{-\boldsymbol{k}n\uparrow}\rangle$, $T^{0}_{\boldsymbol{k}n}=(\langle a_{\boldsymbol{k}n\uparrow}a_{-\boldsymbol{k}n\downarrow}\rangle+\langle a_{\boldsymbol{k}n\downarrow}a_{-\boldsymbol{k}n\uparrow}\rangle)/\sqrt{2}$, and $T^{-1}_{\boldsymbol{k}n}=\langle a_{\boldsymbol{k}n\downarrow}a_{-\boldsymbol{k}n\downarrow}\rangle$. As expected, spin singlet and triplet pairs have even- and odd-parity, respectively.

Note here that the magnitude of on-site pair-amplitude vanishes for the odd-parity triplet pair, while for the singlet case, it depends on the symmetry of Cooper pair. As is well known, the Hund's rule coupling stabilizes the local triplet pair composed of electrons in different orbitals, inevitably leading to the orbital antisymmetric pair with finite on-site amplitude [14]. However, such an even-parity triplet pair does not appear in the weak-coupling limit. In order to make this point clear, it is instructive to consider the on-site pair-amplitude in the two-band model on the Bravais lattice. After simple algebraic calculations, we obtain $\langle c_{\mathbf{i}\alpha\sigma}c_{\mathbf{i}\beta\sigma}\rangle=\sum_{\boldsymbol{k}}\langle a_{\boldsymbol{k}1\sigma}a_{-\boldsymbol{k}2\sigma}\rangle$ for $\alpha\neq\beta$. Namely, the triplet pair induced by the Hund's rule coupling should be composed of electrons on different bands, which is not stable in the weak-coupling limit [15]. We conclude that for the Bravais lattice, odd-parity triplet pair cannot be induced by the Hund's rule coupling.

Next we turn our attention to the pair formation in the non Bravais lattice, typically the honeycomb lattice. For simplicity, we consider only the on-site pair amplitude. Using Eqs. (3) and (6), for even-parity singlet and odd-parity triplet pairs, we obtain

$$\langle\phi^{\mathrm{S}}_{\mathbf{i}1\alpha,\,\mathbf{i}1\beta}\rangle=(1/2)\sum_{n,\boldsymbol{k}}(\Lambda^{\boldsymbol{k}n}_{1\alpha}\Lambda^{-\boldsymbol{k}n}_{1\beta}+\Lambda^{-\boldsymbol{k}n}_{2\alpha}\Lambda^{\boldsymbol{k}n}_{2\beta})S_{\boldsymbol{k}n}, \quad (9)$$

and

$$\langle\phi^{\mathrm{T},z}_{\mathbf{i}1\alpha,\,\mathbf{i}1\beta}\rangle=(1/2)\sum_{n,\boldsymbol{k}}(\Lambda^{\boldsymbol{k}n}_{1\alpha}\Lambda^{-\boldsymbol{k}n}_{1\beta}-\Lambda^{-\boldsymbol{k}n}_{2\alpha}\Lambda^{\boldsymbol{k}n}_{2\beta})T^{z}_{\boldsymbol{k}n}, \quad (10)$$

respectively. Note that in general, $\langle\phi^{\mathrm{T},z}_{\mathbf{i}1\alpha,\,\mathbf{i}1\beta}\rangle$ does *not* vanish. In order to understand this point, it is useful to

remark the relation

$$\langle \phi^{\mathrm{T},z}_{\mathbf{i}1\alpha,\,\mathbf{i}1\beta}\rangle = -\langle \phi^{\mathrm{T},z}_{\mathbf{i}2\alpha,\,\mathbf{i}2\beta}\rangle. \tag{11}$$

The pair satisfying this relation is composed of the local triplet pairs in the unit cell in an out-of-phase manner, as schematically shown in Fig. 2(a). Then, the odd-parity triplet pair induced by the Hund's rule coupling can appear in the non Bravais lattice.

We believe that the above symmetry argument is useful, but it mentions nothing about the stability of the odd-parity pair. For instance, the in-phase combination of local triplet pairs with even-parity [see Fig. 2(b)] seems to dominate the odd-parity pair at a first glance, but the above discussion does not conclude which pair is stabilized. In order to complete the discussion, we cannot avoid to carry out some explicit calculations in an appropriate model. However, if we apply mean-field approximations on the model in which attractive interactions are introduced just by hand, we may lose essential points. Thus, in this paper, we adopt the orbital degenerate Hubbard model, which is widely believed to be a standard model for strongly correlated electron systems. Furthermore, in order to obtain unbiased results, we resort to the numerical method such as exact diagonalization. For the purpose to analyze the paring symmetry in the numerical calculation, we apply the method of the optimization of pair correlation function [14].

Here we take the e_{g}-orbital Hubbard model as [16]

$$\begin{aligned} H &= -\sum_{\langle \mathbf{i}\mu,\mathbf{j}\nu\rangle}\sum_{\sigma,\alpha,\beta} t^{\mathbf{v}}_{\alpha\beta}(c^{\dagger}_{\mathbf{i}\mu\alpha\sigma}c_{\mathbf{j}\nu\beta\sigma} + \mathrm{h.c.}) \\ &+ U\sum_{\mathbf{i},\mu,\alpha}\rho_{\mathbf{i}\mu\alpha\uparrow}\rho_{\mathbf{i}\mu\alpha\downarrow} + U'/2\sum_{\mathbf{i},\mu}\sum_{\alpha\neq\beta}\rho_{\mathbf{i}\mu\alpha}\rho_{\mathbf{i}\mu\beta} \\ &+ J/2\sum_{\mathbf{i},\mu}\sum_{\sigma,\sigma',\alpha\neq\beta} c^{\dagger}_{\mathbf{i}\mu\alpha\sigma}c^{\dagger}_{\mathbf{i}\mu\beta\sigma'}c_{\mathbf{i}\mu\alpha\sigma'}c_{\mathbf{i}\mu\beta\sigma} \\ &+ J'\sum_{\mathbf{i},\mu}\sum_{\alpha\neq\beta} c^{\dagger}_{\mathbf{i}\mu\alpha\uparrow}c^{\dagger}_{\mathbf{i}\mu\alpha\downarrow}c_{\mathbf{i}\mu\beta\downarrow}c_{\mathbf{i}\mu\beta\uparrow}, \end{aligned} \tag{12}$$

where $\langle \mathbf{i}\mu,\mathbf{j}\nu\rangle$ denotes a pair of nearest-neighbor sites, $\mathbf{v}$ indicates a vector connecting the nearest-neighbor sites, α (β) denotes x^2-y^2 ($3z^2-r^2$) orbital, $\rho_{\mathbf{i}\mu\alpha\sigma}=c^{\dagger}_{\mathbf{i}\mu\alpha\sigma}c_{\mathbf{i}\mu\alpha\sigma}$, and $\rho_{\mathbf{i}\mu\alpha}=\sum_{\sigma}\rho_{\mathbf{i}\mu\alpha\sigma}$. In the first term, $t^{\mathbf{v}}_{\alpha\beta}$ is a hopping amplitude depending on orbitals, hopping directions, and lattice type. Hereafter $t^{\mathbf{y}}_{\alpha\alpha}$ along the $\mathbf{y}$ direction (see Fig. 1) is taken as the energy unit. In the Coulomb interaction terms, U, U', J, and J' denote intra-orbital, inter-orbital, Hund's rule, and pair-hopping interactions, respectively. Note the relations of $U=U'+J+J'$ and $J=J'$.

First let us briefly discuss the ground-state phase diagrams for two types of 2D lattices with 8 sites (see Fig. 1). In Fig. 3(a), the phase diagram for the 8-site honeycomb lattice is shown for n=0.75. When J is increased, the FM phase appears, since it can gain the kinetic energy for large J. This is quite general in the orbital degenerate model. For n=1 (not shown here), the FM phase is also found, but the region becomes narrow: It does not touch the line of $J=U'$. For the 2D square lattice with 8 sites, the phase diagram for n=1.5 is quite similar to Fig. 3(a) (see Ref. [14]). For n=1, the phase diagram is also similar to that of the honeycomb lattice with n=1. For n=0.75, the FM phase disappears in the square lattice.

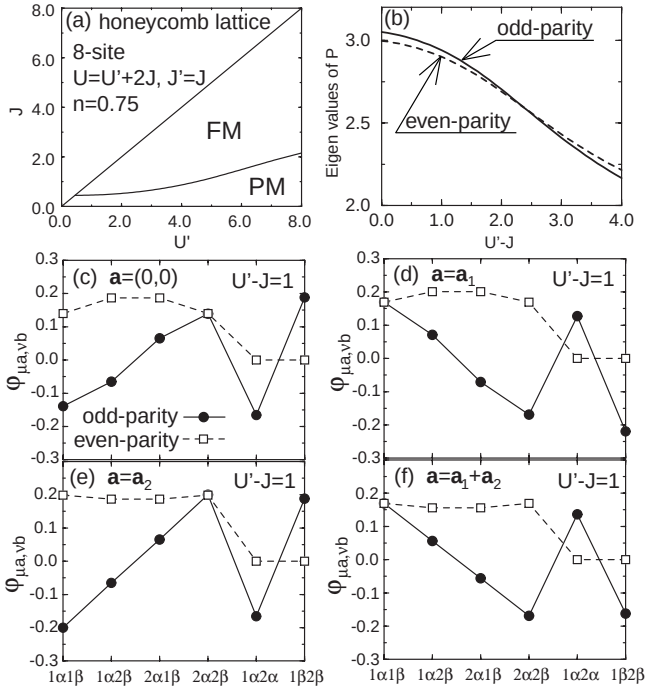

FIG. 3: (a) Ground-state phase diagram for 8-site honeycomb lattice with n=0.75. Note that the region for $J>U'$ is unphysical. (b) Maximum and second maximum eigen-values of P as a function of $U'-J$ in the FM phase. Coefficients of the odd- and even-parity triplet pair correlation functions for (c) $\mathbf{a}$=(0,0), (d) $\mathbf{a}=\mathbf{a}_1$, (e) $\mathbf{a}=\mathbf{a}_2$, and (f) $\mathbf{a}=\mathbf{a}_1+\mathbf{a}_2$, for $U'-J$=1.

Now we focus on the pair in the FM phase to clarify the symmetry of triplet pair induced by the Hund's rule coupling. For the purpose, we measure the triplet pair correlation function, defined as $P=\langle\Phi\Phi^{\dagger}\rangle$, where Φ is a triplet pair operator given by a linear combination of $\phi^{\mathrm{T},z}_{\mathbf{i}\mu\alpha,\,\mathbf{j}\nu\beta}$. The coefficient of each component includes information about pairing symmetry. The pair wavefunction should be determined as the eigen function with the maximum eigenvalue of P [14]. The pairing state thus determined, called "optimized pair", is the most probable candidate for the Cooper pair in actual systems.

Before proceeding to the exhibition of numerical results, let us briefly discuss the meaning of the optimized pair from the conceptual viewpoint of off-diagonal long-range order [13]. In principle, the occurrence of superconductivity is detected when the maximum eigenvalue $\rho_{\max}$ of P becomes the order of N, where N is the number of electrons. In a small cluster calculation, the possi-

ble superconducting pair state should be defined by the eigenstate with $\rho_{\rm max}$, but in order to prove the existence of off-diagonal long-range order, it is necessary to show that $\rho_{\rm max}$ actually becomes the order of N with increasing the system size. However, at some cluster size, a possible pairing state is determined without ambiguity by the eigenstate with $\rho_{\rm max}$.

In Fig. 3(b), we show the eigenvalues of P in the FM phase of the honeycomb lattice for n=0.75. For small U', the eigenstate of P corresponding to $\rho_{\rm max}$ exhibits odd-parity, while the even-parity state appears for large value of U' [17]. As discussed below, the symmetry of those pairing states are consistent with the schematic views shown in Figs. 2(a) and (b). In the fully spin-polarized phase, the relevant interaction is $U'-J$. Then, small $U'-J$ corresponds to the "weak-coupling" region in the FM phase. In that sense, it seems natural that the odd-parity state appears for small $U'-J$, while the even-parity occurs for large $U'-J$.

In order to visualize the paring symmetry, we explicitly express the pair operators as

$$\begin{aligned}\Phi &= \sum_{\mathbf{i},\mathbf{a}}(\varphi^{\mathbf{a}}_{1\alpha,1\beta}c_{\mathbf{i}1\alpha\uparrow}c_{\mathbf{i}+\mathbf{a}1\beta\uparrow}+\varphi^{\mathbf{a}}_{1\alpha,2\beta}c_{\mathbf{i}1\alpha\uparrow}c_{\mathbf{i}+\mathbf{a}2\beta\uparrow}\\ &+ \varphi^{\mathbf{a}}_{2\alpha,1\beta}c_{\mathbf{i}2\alpha\uparrow}c_{\mathbf{i}+\mathbf{a}1\beta\uparrow}+\varphi^{\mathbf{a}}_{2\alpha,2\beta}c_{\mathbf{i}2\alpha\uparrow}c_{\mathbf{i}+\mathbf{a}2\beta\uparrow}\\ &+ \varphi^{\mathbf{a}}_{1\alpha,2\alpha}c_{\mathbf{i}1\alpha\uparrow}c_{\mathbf{i}+\mathbf{a}2\alpha\uparrow}+\varphi^{\mathbf{a}}_{1\beta,2\beta}c_{\mathbf{i}1\beta\uparrow}c_{\mathbf{i}+\mathbf{a}2\beta\uparrow}), \quad (13)\end{aligned}$$

where $\mathbf{a}$ is a vector connecting two unit cells. See Fig. 1(b) for the definitions of $\mathbf{a}$ in the honeycomb lattice.

In Figs. 3(c)-(f), we depict the coefficients $\varphi^{\mathbf{a}}_{\mu\mathrm{a},\nu\mathrm{b}}$ for each $\mathbf{a}$ and $U'-J$=1 [18]. The symmetry argument in the weak-coupling limit has suggested that the odd-parity triplet pair should satisfy the relations, $\varphi^{\mathbf{a}}_{1\alpha,1\beta}=-\varphi^{\mathbf{a}}_{2\alpha,2\beta}$ and $\varphi^{\mathbf{a}}_{1\alpha,2\beta}=-\varphi^{\mathbf{a}}_{2\alpha,1\beta}$ [19]. In Fig. 3(c), these relations are actually satisfied in the pair correlation inside the unit cell for $\mathbf{a}$=$(0,0)$, indicating *unbiased* evidence for the odd-parity triplet pair. As shown in Figs. 3(d)-(f), the above relations for $\mathbf{a}$=(0,0) are also observed for $\mathbf{a}\neq$(0,0) and the pair amplitudes for $\mathbf{a}\neq$(0,0) are in the same order as those for $\mathbf{a}$=(0,0). These results indicate that the triplet pair spatially extends to gain the kinetic energy by keeping the odd-parity symmetry. Here note an in-phase relation in the coefficients for $\mathbf{a}_2$ direction, while an out-of-phase relation for $\mathbf{a}_1$ direction, suggesting a node structure between the unit cells along $\mathbf{a}_1$ direction. Namely, the p-wave-like pair is appearing in this case. Note also another relation $\varphi^{\mathbf{a}}_{1\alpha,2\alpha}\approx-\varphi^{\mathbf{a}}_{1\beta,2\beta}$ for the odd-parity pair, indicating the node of the pair wavefunction in the orbital space.

Finally, we briefly comment on actual compounds from the present result. First, the odd-parity triplet pair in the FM phase induced by the Hund's rule coupling is expected to occur in some f-electron compounds such as UGe_2 and URhGe, although further quantitative calculations are needed based on more realistic models. Second, the triplet pair in ruthenate may not be induced by the local Hund's rule interaction, since the ruthenate is described by the $t_{2\mathrm{g}}$-orbital degenerate Hubbard model on the 2D square lattice. Rather we may consider a scenario based on the pairing induced by the effective interaction among quasi-particles in the Fermi-liquid theory [20].

In summary, we have discussed microscopic aspects of odd-parity triplet pair. The weak-coupling analysis has suggested that the odd-parity triplet pair induced by the Hund's rule coupling does not appear in the Bravais lattice. We have carefully analyzed the triplet pair wavefunction in the FM phase on the 2D square and honeycomb lattices. It has been clarified that the odd-parity triplet pair induced by the local Hund's coupling occurs only for the non Bravais lattice.

The authors are separately supported by the Grant-in-Aid for Scientific Research from Japan Society for the Promotion of Science.

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